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Quantum sensing and metrology for fundamental physics with molecules

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Quantum sensing and metrology use coherent superposition states of quantum systems to detect and measure physical effects of interest. Their sensitivity is typically limited by the standard quantum limit, which bounds the achievable precision in measurements involving nominally identical but uncorrelated quantum systems. Fully quantum metrology involves entanglement in an array of quantum systems, enabling uncertainty reduction below the standard quantum limit. Although ultracold atoms have been widely used for applications such as atomic clocks or gravitational sensors, molecules show higher sensitivity to many interesting phenomena, including the existence of new, symmetry-violating forces mediated by massive particles. Recent advancements in molecular cooling, trapping and control techniques have enabled the use of molecules for quantum sensing and metrology. This Review describes these advancements and explores the potential of the rich internal structure and enhanced coupling strengths of molecules to probe fundamental physics and drive progress in the field.

Ultracold atoms have emerged as a powerful and versatile tool for metrology and sensing applications[1]. For such purposes, atoms typically are prepared in a quantum superposition state, where the relative phase between the states grows at a rate proportional to the physical effect being measured. This phase is then mapped onto a measurable difference in individual state populations (Fig. 1). Slowly moving or trapped ultracold atoms in ultrahigh vacuum are ideal for such measurements, as the phase accumulates over time. This approach is typified by atomic clocks, but also finds applications in detecting various types of external perturbation on the atoms, arising from magnetic and/or electric fields, inertial forces, hypothesized dark matter particles and more.

Atoms can also be used to measure the forces acting within them[1]. For example, ultra-precise measurements of energy levels, when compared with theoretical predictions, can provide insights into quantum electrodynamics, nuclear structure or potential 'fifth' forces, each of which shifts the levels in a characteristic way. Measurements of energy shifts induced by certain configurations of electromagnetic fields can reveal properties of the ordinary weak interaction or uncover exotic new forces that violate charge conjugation–parity (CP) symmetry. These methods commonly achieve precision and sensitivity at a fundamental limit set by quantum mechanics, known as the standard quantum limit (SQL). Here the uncertainty $\delta\varphi$ in a phase φ is given by $\delta\varphi_{\mathrm{SQL}} = 1/(\tau\sqrt{N})$, where τ is the coherence time and N is the number of particles measured.

Molecular sensors can provide enhanced sensitivity to many interesting phenomena by exploiting the additional degrees of freedom in molecules, such as rotations and vibrations[2,3]. For example, the small energy splittings between opposite-parity rotational states amplify the effect of parity-violating interactions by many orders of magnitude[4], and vibrational energy splittings are highly sensitive to fifth forces at the submicrometre range[5,6]. New experimental methods are now enabling the use of cold and ultracold molecules as quantum sensors for such purposes. A crucial requirement for molecule-based sensing

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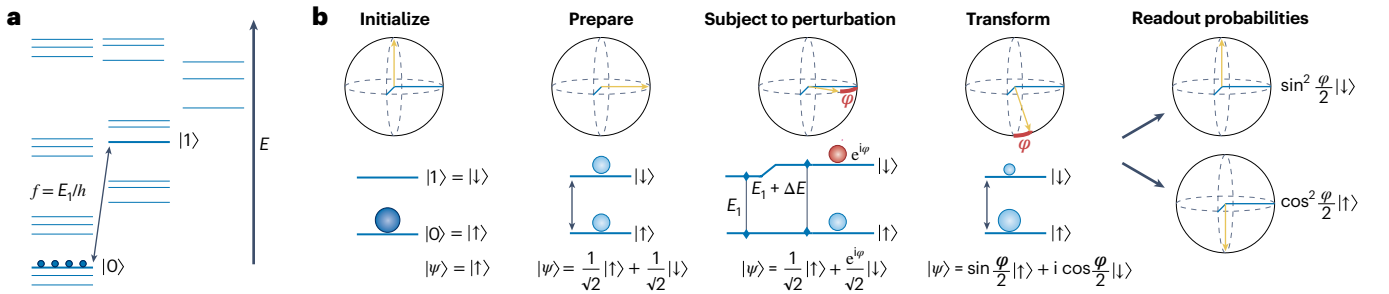

Fig. 1 | A typical quantum sensing protocol with molecules. a, Molecules have a complicated distribution of energy levels, many of which are populated at temperatures ≳1 K. Further cooling can prepare a single quantum state $|0\rangle$ (with energy defined as 0). Application of electromagnetic fields whose frequency f is resonant with the transition from $|0\rangle$ to a single other state $|1\rangle$ (energy E_1), such that $f = E_1/h$ (where h is Planck's constant), creates an effective two-level quantum system useful for sensing. **b**, Any two-level system is mathematically equivalent to a spin-1/2 particle, with eigenstates $|\uparrow\rangle$ and $|\downarrow\rangle$. Any arbitrary state of the system, $|\psi\rangle$, can be written in the form $|\psi\rangle = \cos\theta\,|\uparrow\rangle + e^{i\varphi}\sin\theta\,|\downarrow\rangle$, and visualized to lie on a 'Bloch sphere' at a point with polar coordinates θ (latitude) and φ (longitude). For sensing, the initial state $|0\rangle \equiv |\uparrow\rangle$ is transformed by a resonant pulse into an equal-weight superposition of $|\uparrow\rangle$ and $|\downarrow\rangle \equiv |1\rangle$, after which the perturbation of interest caused by the phenomenon to be sensed leads to a non-zero value $\varphi = \Delta E T/\hbar$, where ΔE is the energy shift and T is the observation time. A second resonant pulse transforms the system to a superposition with relative amplitudes determined by φ, which can be read out by performing state-selective detection of $|\uparrow\rangle$ and/or $|\downarrow\rangle$, with many repetitions to find the average value $\langle\varphi\rangle$ and its uncertainty $\delta\varphi$. This protocol is closely related to work using molecules as qubits[88].

is the cooling of not only external motional degrees of freedom but also internal motions, including vibrations, rotations, and electron and nuclear spins. Cryogenic buffer gas-cooled molecular beams[7] enable modest control over both internal and external molecular motion, and are widely applicable. Direct laser cooling and trapping of molecules, first demonstrated in 2010[8], is now applied to reach submillikelvin temperatures for many diatomic[9] and, recently, polyatomic[10] species. The most exquisite control is obtained with diatomic molecules assembled from ultracold atoms, reaching temperatures in the nanokelvin regime[11–14]. These methods are enabling revolutionary improvements in the ability to create, control and measure long-lived quantum superposition states in molecules[15]. Using particular species and internal degrees of freedom chosen for optimized sensitivity to the effects of interest, molecular systems are becoming the tools of choice for several types of quantum sensing and metrological applications. Here we discuss a few of these applications.

Sensing symmetry-violating forces with molecules

Several cosmological observations, such as the lack of free antimatter in the Universe and the existence of dark matter, suggest the presence of undiscovered fundamental particles and new forces mediated by them. Such forces are often expected to violate symmetries such as CP. For example, new CP-violating (CPV) interactions are needed in many models to explain the matter–antimatter asymmetry[16], as CP-preserving effects cannot bias the production of matter over antimatter in the early Universe. In addition, theories that introduce new particles beyond those in the standard model typically open new mechanisms for CP violation, making the search for such effects a fairly generic way to search for physics beyond the standard model.

New forces can lead to observable effects in ordinary matter, associated with virtual exchange of such particles. Typically, the size of the observable is proportional to the inverse square of the new particle's mass. Methods to detect symmetry-violating interactions can be exquisitely sensitive, as they are clearly distinguishable from the dominant strong and electromagnetic forces. Hence, experiments of this type can probe the existence of extremely massive new particles. However, using any given molecular species requires detailed theoretical analysis to quantify its sensitivity to fundamental symmetry violations, and extensive spectroscopic study to understand its energy-level structure. Rapid advances in these areas are key to enabling molecular searches for symmetry-violating effects.

CPV effects in atoms and molecules

The effects of CPV interactions often manifest through the existence of particular distributions of charges and currents linked to the spin axis of a quantized particle, such as an electric dipole moment (EDM) or other electromagnetic (EM) moments. In a molecule or atom, such EM moments of an electron or a nucleus interact with the EM fields produced by other charged particles in the system. Such EM interactions are strongest when an electron (e) is in close proximity to or even inside a nucleus ($\mathcal{N}$). The largest e–$\mathcal{N}$ overlaps occur when electrons are in s orbitals around heavy nuclei with high atomic number ($Z \gg 1$), as these nuclei also produce the largest EM fields. The same conditions also maximize the effect of CPV e–$\mathcal{N}$ interactions, which can be described as short-range 'contact' e–$\mathcal{N}$ interactions when mediated by massive particles.

Although an electron experiences strong EM fields near a heavy nucleus, the vector average of these fields, as seen by an electron in an s orbital, vanishes. To create a net average EM field, the electron must spend more time on one side of the nucleus than the other. The same condition holds for activating CPV e–$\mathcal{N}$ interactions. This can be accomplished by polarizing the electron orbital, using a laboratory electric (E) field. Electron orbitals in atoms require roughly one atomic unit of E field, ~5 GV cm^{-1}, to be fully polarized. However, as static lab E fields typically cannot exceed ~200 kV cm^{-1}, the polarization is, at best, tiny. Consequently, in atoms, the net average EM field near the nucleus is orders of magnitude smaller than its naive value.

By contrast, in polar molecules, the strong internal E field caused by the other nucleus can fully polarize an electron along the molecular axis, into an s–p hybridized (σ) orbital. In this case, polarizing only the rotational motion of the molecule is sufficient for the electron to experience the largest possible net average EM fields (or CPV contact interactions). As the rotation of typical polar molecules can be fully polarized in lab-scale E fields (~30 kV cm^{-1} or, often, much smaller), the net size of CPV effects in molecules can be amplified by many orders of magnitude relative to atoms. The recent development of new methods to create heavy molecular species and to prepare, control and measure their quantum states is now making it possible to take advantage of this huge amplification.

Electron EDM

The most notable success in using molecules as quantum sensors has been in searches for a CPV EDM along the spin of the electron (Fig. 2). Although these experiments have not yet detected the electron EDM,

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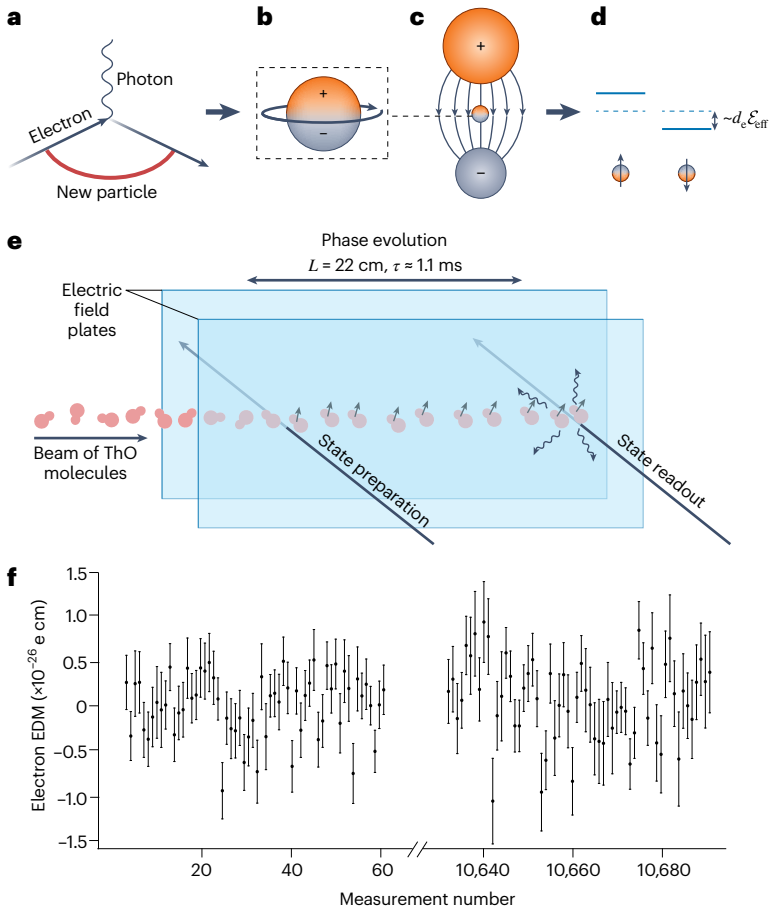

Fig. 2 | CP violation effects. a,**b**, New particles mediating symmetry-violating forces can couple to the electron (**a**), giving rise to a permanent electron EDM, d_e, along its spin axis (**b**). **c**,**d**, The valence electron in a polar molecule (**c**) can experience very large effective electric fields $\mathcal{E}_{eff}$, giving rise to molecular energy shifts ~ $d_e\mathcal{E}_{eff}$ (**d**). **e**, Schematic of an example experiment, ACME[18]. A beam of ThO molecules is sent through lasers that prepare a coherent superposition (Fig. 1), which evolves as the molecules travel through a region of length L in the presence of electric and magnetic fields, and is then read out by different lasers. **f**, Many repeated measurements[89] give enough statistical sensitivity to probe the electron EDM with sufficient precision to search for new fundamental particles and forces. Panel **f** adapted with permission from ref. 89 under a Creative Commons licence CC BY 3.0.

their sensitivity has advanced substantially in the past decade[17–19]. They are currently sensitive to the existence of certain new particles with mass at the 10–50 TeV scale, indicating that such particles must have even higher mass, well beyond the reach of direct searches using the Large Hadron Collider. Although the standard model of particle physics predicts a non-zero electron EDM, experimental sensitivity will have to improve by roughly a million-fold to detect it at the predicted size[20]. By contrast, in many theories of physics beyond the standard model, the new particles can lead to a much larger electron EDM, even if they have much higher masses than standard model particles. As molecular EDM experiments appear poised to probe even higher mass scales soon, we discuss this topic in some detail.

The electron EDM leads to a negative (positive) energy shift when the electron spin, and hence the EDM, is aligned (anti-aligned) with an electric field. As described above, an electron experiences the strongest field when in a σ molecular orbital around a highly charged nucleus, and when the rotation of the molecule is fully polarized. Consider, for example, an electron in the thorium monoxide molecule (ThO). Here, owing to the heavy nucleus Th (Z = 90), the electron EDM experiences a net effective E field of ~80 GV cm^{-1} (refs. 21,22) when the molecular rotation is polarized with a modest laboratory electric field (~10 V cm^{-1} or more).

The first molecular experiment to set a strict limit on the electron EDM used a free-radical species (YbF) with one valence σ electron[23]. More recent experiments, such as ACME[17,18] (the Advanced Cold Molecule electron EDM experiment; see below) and one using trapped ions[19], use molecules (ThO, HfF^+) with a second valence electron in a different molecular orbital. This serves to cancel the magnetic moment of the electron in the EDM-sensitive σ orbital, which reduces both noise and possible systematic errors due to stray magnetic fields. It also imbues the molecule with pairs of quantum states that have extremely high polarizabilities of opposite sign (due to the phenomenon known as Λ- or Ω-doubling[24]). The energy shift associated with the EDM is of opposite sign in these pairs of states, allowing for the suppression of many possible systematic errors by measuring in both states[25].

Existing experiments continue to rapidly improve. The experiment using HfF^+, performed at JILA (Boulder, CO, USA), recently reported an upper bound on the electron EDM a factor of 2.5 times smaller[19] than the previous best limit from ACME set in 2018[18]. Both experiments had statistical uncertainty near the SQL. Systematic errors were even smaller, and could be reduced further with better statistics. Both experiments are currently undergoing upgrades to increase the useful number of molecules and the coherent spin evolution time, among other improvements. ACME projects a factor of ~30 increased sensitivity relative to its best previous result[26,27] and the JILA experiment is aiming for similar ultimate sensitivity. This will explore mass ranges of ~150 TeV in typical theories of physics beyond the standard model. Concurrently, new approaches are also being developed to reach sensitivity orders of

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BOX 1

New approaches for molecular EDM searches

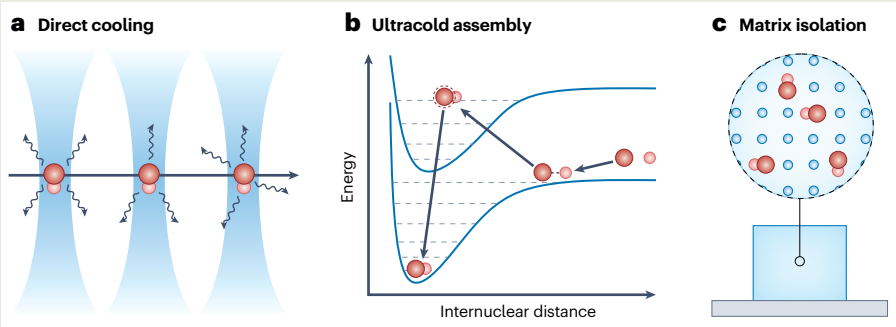
Here we describe some promising avenues that are emerging for the improvement of sensitivity of molecular-based EDM sensors.

Direct cooling. Laser-cooled and trapped neutral molecules[9] offer a route to large count rates with advanced quantum control, as demonstrated by the success of atomic clocks based on ultracold atoms. Whereas most experiments use diatomic species[90], polyatomic species are also amenable to laser cooling, and provide additional capabilities to engineer structure and electromagnetic properties to enhance EDM searches, such as high polarizability and tunable magnetic moments[91–93]. Many promising species are being explored[29,30], including BaF, YbF, RaF, SrOH, YbOH and RaOH.

Ultracold assembly. Ultracold, trapped molecules can be formed by coherent assembly from ultracold, trapped atoms, thus offloading the difficulty of cooling to the simpler case of atoms[11]. The challenge is to find species that are heavy and polar, as assembled molecules tend to be covalently bonded. However, molecules containing silver atoms, such as FrAg and RaAg, are highly polar and can probably be assembled at ultracold temperatures[94,95].

Matrix isolation. While gas-phase molecules offer the maximum possible coherence, those in the solid state can offer unrivalled particle numbers. There has been a recent resurgence in EDM searches using polar molecules isolated in crystals[96–98] or noble gas matrices[99,100]. Such optically transparent media enable methods for quantum control and readout, and show potentially high sensitivity if systematic errors due to the complex environment of a solid material can be controlled.

Each of these methods offers a route to improve electron-EDM sensitivity by several orders of magnitude. For example, an experiment with 10^6 optimally sensitive molecules, 100 s coherence time and 1 year of integration could detect an electron EDM, d_e, with magnitude as small as $|d_e| < 10^{-33}$ e cm, thereby probing energy scales up to around 8,000 TeV, far beyond the reach of any conceivable future high-energy collider.



New methods for electron EDM searches. Diatomic molecules for EDM searches typically consist of a heavy atom of a metallic element (red, larger spheres) bound to a lighter, electronegative atom (pink, smaller spheres). **a**, Certain molecules can scatter many photons; such species can be laser-cooled directly, then loaded, for example, into a tweezer array, where they can be imaged. **b**, Certain pairs of ultracold atoms can be assembled into a bound molecule, for example, through magnetoassociation to a weakly bound state followed by coherent optical transfer through excited bound electronic states. **c**, Molecules can be embedded in cryogenic solids and oriented by the lattice.

magnitude better than at present, on the ~10-year timescale. Some of these efforts are discussed in Box 1.

CPV nuclear moments

The nuclei in molecules also provide interesting opportunities for detecting new CPV forces, but with sensitivity to different underlying physics compared with the electron EDM, namely, forces coupling to quarks, gluons, nucleons, pions and so on. Although nuclear EDMs are shielded by surrounding electrons, CPV interactions can manifest themselves through a nuclear Schiff moment (NSM; a dipole-like charge distribution on the nuclear surface) or/and a magnetic quadrupole moment (MQM; a current distribution like a pair of nearby coils with currents flowing in opposite directions). Similar to the electron EDM, signals from these effects are greatly amplified—typically ~1,000-fold—in heavy molecules compared with in atoms. A molecular NSM search was completed in TlF molecules approximately 30 years ago[28]. Several new, similar experiments are actively being developed[29,30] using modern molecular techniques with TlF[31] and Ra-containing neutrals and ions.

In certain nuclei, the NSM or MQM itself can be greatly amplified when the nucleus has a deformed shape. In particular, quadrupolar or octupolar deformations amplify the MQM and NSM, respectively. Certain heavy, radioactive isotopes, such as ^{223}Fr, ^{225}Ra, ^{227}Ac, ^{227}Th and ^{229}Pa, offer typically ~1,000-fold enhancement of NSMs, for the same underlying source of CPV, due to such deformations[30,32]. MQMs of many stable deformed isotopes can yield CPV energy shifts ~10 times larger than from ordinary NSMs[33,34]. No molecular MQM search has been completed, but several are underway or under development[29,30], using ^{173}YbF, ^{173}YbOH, ^{181}TaO$^+$, ^{229}ThF$^+$ and ^{175}LuOH$^+$.

Electroweak interactions

The parity-violating electroweak interaction (EWI) leads to energy shifts in atoms and molecules placed in a left- or right-handed configuration of electromagnetic fields. Measurements of these shifts, combined with accurate calculations of atomic and molecular structure, can provide information on aspects of the EWI inaccessible to other methods. In molecules, one aspect of the EWI, which couples an electron to the spin of a nucleus, can be amplified by orders of magnitude relative to atoms by pushing opposite-parity rotational levels to near degeneracy using the Zeeman effect[35]. In heavy molecules, this coupling comes primarily from EWIs between protons and neutrons in the nucleus, leading to a toroidal current distribution in the nucleus known as a nuclear anapole moment (NAM), which interacts magnetically with

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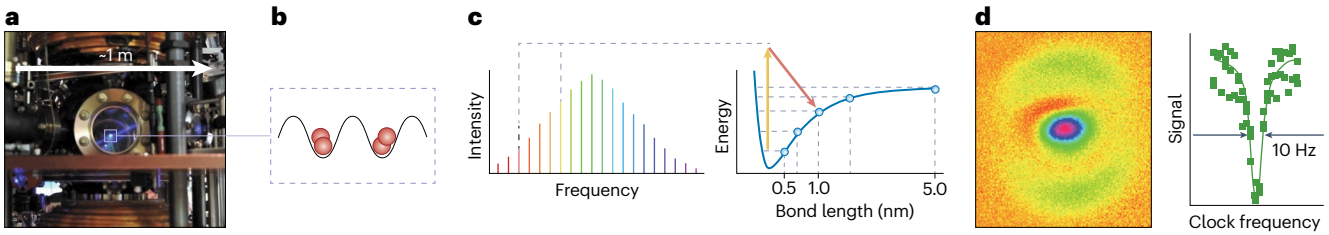

Fig. 3 | Molecular clock set-up. Molecular clocks can be based on trapped ionic or ultracold neutral molecules[40]. **a**,**b**, Here, the vacuum chamber (**a**) contains a cloud of ultracold atoms that are assembled into ultracold diatomic molecules trapped in an optical lattice (**b**). **c**, The vibrational energies of these molecules are probed using two-colour light precisely calibrated and stabilized with a frequency comb. **d**, Molecules remaining in the original state are imaged to obtain narrow clock spectra[40]. Panel **a** courtesy of Tanya Zelevinsky's lab. Panel **d** adapted with permission from ref. 40 under a Creative Commons licence CC BY 4.0.

the spin of penetrating molecular electrons. Measurements of NAMs can provide new information on these intranuclear EWIs, which remain poorly understood. A proof-of-principle experiment recently demonstrated a method to measure NAMs using diatomic molecules, yielding sensitivity well beyond the previous state of the art using atoms[36]. This method can be applied to measure NAMs of isotopes of many different elements[35]. Applying similar techniques with laser-cooled and trapped neutral[37] or ionic[35,38] molecules will probably enable measurements of even smaller EWI effects, such as the direct couplings of Z^0 bosons to neutron or proton spins.

Molecular metrology and applications in fundamental physics

Molecular clocks

Molecules present enticing opportunities in sensing of fundamental phenomena that have proven to be elusive with other methods. In particular, high-precision state control of molecules enables the development of clocks that can explore physics from a complementary angle to atomic clocks. Atomic clocks are currently the ultimate sensors of space–time, with sensitivity to gravitational redshifts approaching the microscopic scale[39]. Molecular clocks, with their rotational and vibrational dynamics, present an interesting alternative approach.

Molecular clocks (Fig. 3) can utilize assembled ultracold neutral molecules such as Sr_2 (ref. 40), laser-cooled neutral radicals such as CaF (ref. 41) or molecular ions such as N_2^+ (ref. 42). These systems feature narrow transitions, state-insensitive trapping options and low sensitivities to external fields. Their advantages for space–time sensing resolution over atomic clocks could include longer natural lifetimes and lower field sensitivity. However, lower densities and less well-developed state-insensitive trapping are current challenges for the development of molecular clocks. Terahertz frequencies of molecular clocks based on vibrational transitions are unlikely to be competitive with atomic clocks for absolute sensitivity. Nonetheless, they offer benefit for metrology in a new spectral region where secondary frequency standards are scarce[43], and where there is a need for continued technological developments that will in turn enhance studies of molecular and solid-state systems. A complete evaluation of systematic effects for an Sr_2 clock yielded a fractional uncertainty of $<5 \times 10^{-14}$ (ref. 40), a promising starting point for high-precision sensing of the types discussed in the remainder of this section.

Dark matter detection

Several observed cosmological phenomena could be explained via hypothetical dark matter particles[44]. One proposed type of dark matter consists of light (mass $\lesssim 10^{-1}$ eV c^{-2}) bosonic particles that form a coherent field and interact with ordinary particles not only gravitationally but also via very weak additional forces. For example, spin-zero particles known as moduli—which arise naturally in string theories—can have couplings that modify the masses of ordinary particles and the strength of known forces proportional to the amplitude of these new fields. As masses and force strengths are usually thought of as eternally fixed, changes in the new fields' amplitudes would appear as variations in some fundamental constants, on a timescale inversely proportional to the dark boson mass[45]. Molecule experiments are currently most sensitive to very light dark matter candidates (mass $<10^{-15}$ eV c^{-2}). Molecular vibrational transitions depend directly on the electron-to-proton mass ratio η, and hence are suitable for characterizing possible dynamics of η (ref. 42). Atomic clocks based on hyperfine transitions also are proportional to η but depend on a nuclear magnetic g-factor as well. Comparisons of laboratory molecular spectra with observations from the early Universe constrain η drifts to ~10^{-7} over 10^{10} years, whereas the limit from atomic hyperfine clocks is at a similar level of ~4×10^{-17} per year[46] (assuming a constant g-factor). The best laboratory molecular limits on η variations are obtained using a clock transition in ultracold, untrapped KRb molecules[47], though at a level ~300 times less sensitive than from atomic clocks. However, by using much higher-sensitivity vibrational transitions in deeply bound non-polar molecules such as O_2^+, molecular clock experiments could surpass the sensitivity of other methods[42].

Fundamental constants, quantum electrodynamics and fifth forces

Comparing molecular clocks with state-of-the-art calculations of their transition frequencies enables the use of molecules as quantum sensors for detecting fine details of known forces or possible new physical forces. Measurements of rotational–vibrational transitions in hydrogen molecules (H_2, H_2^+ and their isotopologues), described by quantum electrodynamics (QED) and using as inputs only the fine structure constant α, η and the Rydberg constant R_∞, provide a paradigmatic example of this approach. Spectroscopy of H_2^+ is a competitive means to measure the absolute size of η (refs. 48,49), whereas the strictest test of molecular QED was obtained with HD^+ (ref. 50). Rapid developments in molecular quantum control are inspiring concurrent efforts in high-precision QED calculations for heavy molecules, an endeavour traditionally limited to few-body systems such as H_2.

This type of comparison between measurements and theory also constrains hypothetical new fifth forces. Diatomic molecules are particularly sensitive to nucleon–nucleon fifth forces that are absent in atoms. Such forces could include short-range gravity that does not follow Newton's law, or forces mediated by virtual exchange of moduli. Precise spectroscopy and calculations with HD^+ limited such forces to ~10^{-11} of the electromagnetic interaction strength at the ångström scale, corresponding to moduli with ~keV c^{-2} mass[50]. Although direct tests performed with cold neutron beams have so far provided constraints ~300 times stronger[51], molecular-based methods offer substantial room for improvement. For example, weakly bound Yb_2 molecules have been used already for proof-of-principle constraints on non-Newtonian gravity at ~5-nm-length scales[6]. Deeply bound Sr_2 molecules[40] feature smaller bond lengths of ~0.5 nm and have the potential to improve the

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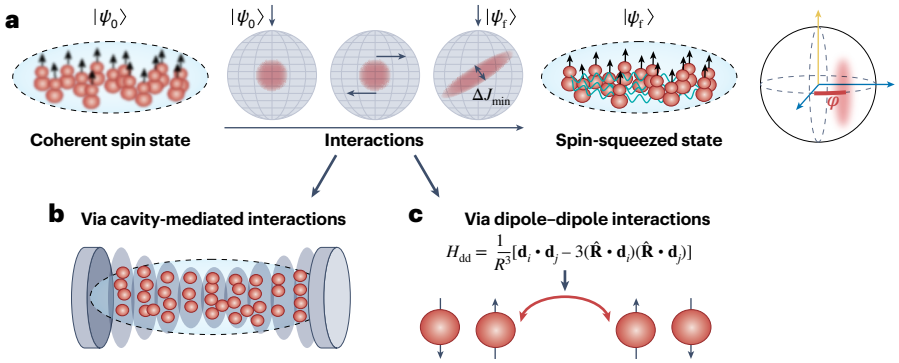

Fig. 4 | Quantum-enhanced sensing. a–c Spin-squeezed states are *N*-particle entangled states that can achieve quantum-enhanced sensitivity and bandwidth compared with what is possible with uncorrelated states. (**a**), A spin-squeezed state of *N* two-level (pseudo spin-1/2) systems can be prepared by starting with a coherent state of the collective pseudo-spin, $|\psi_0\rangle$ (represented as a blurry image to illustrate its limited sensitivity), then letting it evolve under targeted inter-particle interactions. After evolution under such entangling interactions, the coherent spin state can be transformed into a spin squeezed state, $|\psi_f\rangle$. In such a state, the variance of the collective spin projection along one specific direction perpendicular to the mean spin can be reduced to a value $(\Delta J)^2_{\mathrm{min}}$ that is smaller than $N/4$, and as small as 1/4. As the individual pseudo-spins accumulate phase φ, the collective spin vector traces an arc of length $s = J\varphi$ on the collective Bloch sphere. The uncertainty in this length, $\delta s = \Delta J_{\mathrm{min}}$, corresponds to a phase uncertainty $\delta\varphi = \Delta J_{\mathrm{min}}/J$ that is smaller than $\delta\varphi_{\mathrm{SQL}}$ and as small as $\delta\varphi_{\mathrm{H}}$. The interactions required for squeezing can be provided in various ways, such as cavity-mediated interactions (**b**) or electric dipole–dipole (**c**) interactions. The latter are particularly appealing for use with molecular systems as they are naturally present between rotational states of polar molecules. The dipolar interactions between two particles, i,j, with dipole moments $\mathbf{d}_{i,j}$ respectively, can be described by the Hamiltonian H_{dd}. As a function of the distance vector **R** between the particles, H_{dd} scales as R^{-3}; it also displays anisotropic interactions that depend on the orientation of $\mathbf{d}_{i,j}$ relative to each other, and to **R**. Both of these interactions enjoy electromagnetic control and can be enriched by controlling nuclear, spin, vibrational and electronic degrees of freedom in molecules.

best current bounds on new forces around this scale, particularly by comparing molecular isotope shifts with theory[52].

Towards fully quantum metrology with molecules

The experimental methods required to perform the type of measurements discussed so far are often complex, and can provide only moderate numbers of particles *N*. This, in turn, limits the precision of phase measurements at the SQL, when the quantum states of the *N* particles are uncorrelated. However, measurements using many-particle entangled states can enable greater precision, with the ultimate limit set by the Heisenberg uncertainty $\delta\varphi_{\mathrm{H}} = \delta\varphi_{\mathrm{SQL}}/\sqrt{N}$ (ref. 53). Hence, to take advantage of the full potential offered by molecules for sensing, it is crucial to generate robust entangled states that maximize sensitivity for the limited particle numbers available in experiments.

In ensembles of *N* two-level systems (that is, psuedo spin-1/2 systems), it is useful to consider the collective psuedo-spin, $J = N/2$, and to visualize its states on a collective Bloch sphere of radius *N*. If all individual pseudo-spins are aligned along a given direction in their individual Bloch spheres, this forms a so-called spin coherent state of the collective pseudo-spin (Fig. 4a). In such states, the *N* individual systems are uncorrelated, and the variance $(\Delta J)^2$ of the collective spin along any direction perpendicular to the mean spin is equal to $N/4$.

Among the simplest entangled states that can give a metrological advantage over uncorrelated particles are the so-called spin-squeezed states[53–55]. In these states, quantum correlations between the particles help to partially cancel quantum projection noise of the collective spin, thus enabling better angular resolution of rotations of the collective spin along one targeted direction, at the expense of increasing the noise along another angular direction that is irrelevant for the experiment[56]. For example, if the mean collective spin points along the *x* direction of the Bloch sphere describing the *N*-particle collective pseudo-spin, and the state is spin-squeezed in the *y* direction, then it can be used to estimate the accumulated phase from rotations around the *z* axis with sensitivity below the SQL (Fig. 4a).

A paradigmatic strategy to create this form of entangled state is through the dynamical generation of all-to-all, state-dependent Ising interactions. If we characterize the collective pseudo-spin $J = N/2$ of an ensemble of *N* atoms by its total projection *M* along a quantization axis with the state $|J = N/2, M\rangle$, the all-to-all Ising Hamiltonian has eigenvalues proportional to M^2. These types of interactions dynamically shear the classical noise distribution of an initial *N*-particle spin coherent state, with all spins initially polarized along the equator of their individual Bloch spheres, into a spin-squeezed state. Although all-to-all interactions have been achieved in atomic arrays via photon-mediated interactions in optical cavities[57–60] or through collisional interactions in Bose–Einstein condensates[61], achieving them in ultracold-molecule arrays remains an outstanding challenge. Nevertheless, further developments in optical and microwave cavities (Fig. 4b) could open a path for the generation of photon-mediated interactions for spin squeezing.

A more direct route is to generate spin-squeezed states by harnessing the natural interactions between molecules (Fig. 4c), without the need of additional cavities. In recent years, it has been theoretically suggested[62,63] that dipolar interactions, such as those present in arrays of polar molecules[64–68], can indeed give rise to spin-squeezed states with similar metrological utility as the ones obtained by all-to-all interactions. Several experiments have already started to provide validation of such claims[69–72], offering encouraging prospects.

Initial theoretical investigations of squeezing based on electric dipole–dipole interactions between molecules focused on unity-filled arrays of molecules trapped in either deep optical lattices or optical tweezer arrays, where motional degrees of freedom are frozen and on-site collisions suppressed[73,74]. While these conditions are ideal for entanglement generation, they can be hard to achieve experimentally. More recently, mobile molecules confined in stacks of two-dimensional pancakes[63,75] were suggested as a promising alternative for entanglement generation at much simpler conditions and even in dilute samples. Furthermore, the possibility to tune molecular dipolar interactions through the manipulation of nuclear and/or electron spin levels[73,76–78] can offer additional control knobs to enhance coherence times. This could facilitate entanglement generation and detection in a broad class of molecular systems.

We note that the generation of spin squeezing is totally compatible with the Ramsey spectroscopic protocol described in Fig. 1b. The spin-squeezing generation is just an additional step that takes place between the 'prepare' and 'subject to perturbation' steps in Fig. 1b. It is desirable that the interactions relevant for the spin-squeezing generation are present during only the squeezing generation step. To accomplish this task, one can use an auxiliary molecular state that couples to the $|\downarrow\rangle$ state by absorbing a cavity photon, or via dipolar mediated interactions. After the spin-squeezing generation is achieved, one needs only to coherently transfer the atoms in this auxiliary state to the desired $|\uparrow\rangle$ state that is sensitive to the target signal. A scheme for applying such a protocol to a molecular electron-EDM measurement was proposed recently[79].

Outlook

Although quantum sensing and metrology with molecules have already provided substantial contributions in searches for the electron EDM, the field is still in its infancy. However, rapid advances in methods to cool both centre-of-mass and internal motions are enabling experiments with full quantum control over an increasing range of molecular species, opening the door to optimized sensitivity in new and different applications. One striking example is the emerging use of heavy, radioactive molecules[80] containing octupole-deformed nuclei. Experiments under development using ultracold, trapped molecules containing such isotopes promise ~1,000-fold improved statistical sensitivity to new CPV hadronic physics[30], although a thorough analysis of possible systematic errors in such systems remains to be performed. These experiments appear especially amenable to the benefits of spin squeezing, which seems likely to be demonstrated in molecules in the next few years. Overall, the forthcoming advances in molecular experiments probing CPV in electrons, hadrons and e–$\mathcal{N}$ interactions promise to serve as a powerful tool for discovery of new fundamental physics in the next couple of decades.

Although not exploiting quantum superpositions, cooling of molecular internal degrees of freedom is enabling various new sensing applications. The ability to cool fairly complex species[81] has opened a new way to sense molecular chirality[82]. It also promises to enable new approaches to thermometry[83] and dark matter detection[84], by observing resonant excitations due to blackbody radiation or light quantum fields, respectively.

Moreover, cryogenic cooling has already made it possible to demonstrate quantum control over molecular chirality[85]. Further developments in this area, such as the possible laser cooling of chiral molecules[86], promise to open a path to finally detecting the long-anticipated energy difference between different enantiomers of a chiral molecule due to the EWI[87]. As the field of ultracold molecules continues to advance, further ideas for harnessing the rich quantum resources associated with molecular degrees of freedom are likely to emerge, making this a most exciting time for the field.

References

1. Safronova, M. S. et al. Search for new physics with atoms and molecules. *Rev. Mod. Phys.* **90**, 025008 (2018).
2. DeMille, D. Diatomic molecules, a window onto fundamental physics. *Phys. Today* **68**, 34–40 (2015).
3. Hutzler, N. R. Polyatomic molecules as quantum sensors for fundamental physics. *Quantum Sci. Technol.* **5**, 044011 (2020).
4. Sushkov, P. & Flarnbaum, V. Parity breaking effects in diatomic molecules. *Zh. Eksp. Teor. Fiz.* **75**, 1208–1213 (1978).
5. Salumbides, E. J. et al. Bounds on fifth forces from precision measurements on molecules. *Phys. Rev. D* **87**, 112008 (2013).
6. Borkowski, M. et al. Weakly bound molecules as sensors of new gravitylike forces. *Sci. Rep.* **9**, 14807 (2019).
7. Hutzler, N. R., Lu, H.-I. & Doyle, J. M. The buffer gas beam: an intense, cold, and slow source for atoms and molecules. *Chem. Rev.* **112**, 4803–4827 (2012).
8. Shuman, E. S., Barry, J. F. & DeMille, D. Laser cooling of a diatomic molecule. *Nature* **467**, 820–823 (2010).
9. Fitch, N. J. & Tarbutt, M. R. Laser-cooled molecules. *Adv. Mol. Opt. Phys.* **70**, 157–262 (2021).
10. Augenbraun, B. L. et al. Direct laser cooling of polyatomic molecules. *Adv. At. Mol. Opt. Phys.* **72**, 89–182 (2023).
11. Bohn, J. L., Rey, A. M. & Ye, J. Cold molecules: progress in quantum engineering of chemistry and quantum matter. *Science* **357**, 1002–1010 (2017).
12. De Marco, L. et al. A degenerate Fermi gas of polar molecules. *Science* **363**, 853–856 (2019).
13. Schindewolf, A. et al. Evaporation of microwave-shielded polar molecules to quantum degeneracy. *Nature* **607**, 677–681 (2022).
14. Bigagli, N. et al. Observation of Bose–Einstein condensation of dipolar molecules. Preprint at https://arxiv.org/abs/2302.10161 (2023).
15. Langen, T., Valtolina, G., Wang, D. & Ye, J. Quantum state manipulation and cooling of ultracold molecules. *Nat. Phys.* https://doi.org/10.1038/s41567-024-02423-1 (2024).
16. Dine, M. & Kusenko, A. Origin of the matter–antimatter asymmetry. *Rev. Mod. Phys.* **76**, 1–30 (2003).
17. Baron, J. et al. Order of magnitude smaller limit on the electric dipole moment of the electron. *Science* **343**, 269–272 (2014).
18. Andreev, V. et al. Improved limit on the electric dipole moment of the electron. *Nature* **562**, 355–360 (2018).
19. Roussy, T. S. et al. An improved bound on the electron's electric dipole moment. *Science* **381**, 46–50 (2023).
20. Ema, Y., Gao, T. & Pospelov, M. Standard model prediction for paramagnetic electric dipole moments. *Phys. Rev. Lett.* **129**, 231801 (2022).
21. Fleig, T. & Nayak, M. K. Electron electric dipole moment and hyperfine interaction constants for ThO. *J. Mol. Spectrosc.* **300**, 16–21 (2014).
22. Skripnikov, L. V. Combined 4-component and relativistic pseudopotential study of ThO for the electron electric dipole moment search. *J. Chem. Phys.* **145**, 214301 (2016).
23. Hudson, J. J. et al. Improved measurement of the shape of the electron. *Nature* **473**, 493–496 (2011).
24. Brown, J. M. & Carrington, A. *Rotational Spectroscopy of Diatomic Molecules* (Cambridge Univ. Press, 2003); https://doi.org/10.1017/CBO9780511814808
25. DeMille, D. et al. Search for the electric dipole moment of the electron using metastable PbO. *AIP Conf. Proc.* **596**, 72–83 (2001).
26. Wu, X. et al. Electrostatic focusing of cold and heavy molecules for the ACME electron EDM search. *New J. Phys.* **24**, 073043 (2022).
27. Ang, D. G. et al. Measurement of the H $^3\Delta_1$ radiative lifetime in ThO. *Phys. Rev. A* **106**, 022808 (2022).
28. Cho, D., Sangster, K. & Hinds, E. A. Search for time-reversal-symmetry violation in thallium fluoride using a jet source. *Phys. Rev. A* **44**, 2783–2799 (1991).
29. Alarcon, R. et al. Electric dipole moments and the search for new physics. Preprint at https://arxiv.org/abs/2203.08103 (2022).
30. Arrowsmith-Kron, G. et al. Opportunities for fundamental physics research with radioactive molecules. *Rep. Prog. Phys.* https://doi.org/10.1088/1361-6633/ad1e39 (2023).
31. O Grasdijk, O. et al. CeNTREX: a new search for time-reversal symmetry violation in the ^{205}Tl nucleus. *Quantum Sci. Technol.* **6**, 044007 (2021).
32. Auerbach, N., Flambaum, V. V. & Spevak, V. Collective T- and P-odd electromagnetic moments in nuclei with octupole deformations. *Phys. Rev. Lett.* **76**, 4316–4319 (1996).

33. Flambaum, V. Spin hedgehog and collective magnetic quadrupole moments induced by parity and time invariance violating interaction. *Phys. Lett. B* **320**, 211–215 (1994).
34. Flambaum, V., DeMille, D. & Kozlov, M. Time-reversal symmetry violation in molecules induced by nuclear magnetic quadrupole moments. *Phys. Rev. Lett.* **113**, 103003 (2014).
35. DeMille, D., Cahn, S. B., Murphree, D., Rahmlow, D. A. & Kozlov, M. G. Using molecules to measure nuclear spin-dependent parity violation. *Phys. Rev. Lett.* **100**, 023003 (2008).
36. Altuntaş, E., Ammon, J., Cahn, S. B. & DeMille, D. Demonstration of a sensitive method to measure nuclear-spin-dependent parity violation. *Phys. Rev. Lett.* **120**, 142501 (2018).
37. Norrgard, E. B. et al. Nuclear-spin dependent parity violation in optically trapped polyatomic molecules. *Commun. Phys.* **2**, 77 (2019).
38. Karthein, J. et al. Electroweak nuclear properties from single molecular ions in a Penning trap. Preprint at https://arxiv.org/abs/2310.11192 (2023).
39. Bothwell, T. et al. Resolving the gravitational redshift across a millimetre-scale atomic sample. *Nature* **602**, 420–424 (2022).
40. Leung, K. H. et al. Terahertz vibrational molecular clock with systematic uncertainty at the 10^{-14} level. *Phys. Rev. X* **13**, 011047 (2023).
41. Barontini, G. et al. Measuring the stability of fundamental constants with a network of clocks. *EPJ Quantum Technol.* **9**, 12 (2022).
42. Hanneke, D., Kuzhan, B. & Lunstad, A. Optical clocks based on molecular vibrations as probes of variation of the proton-to-electron mass ratio. *Quantum Sci. Technol.* **6**, 014005 (2021).
43. Riehle, F., Gill, P., Arias, F. & Robertsson, L. The CIPM list of recommended frequency standard values: guidelines and procedures. *Metrologia* **55**, 188 (2018).
44. Bertone, G. & Hooper, D. History of dark matter. *Rev. Mod. Phys.* **90**, 045002 (2018).
45. Uzan, J.-P. Varying constants, gravitation and cosmology. *Living Rev. Relativ.* **14**, 2 (2011).
46. Lange, R. et al. Improved limits for violations of local position invariance from atomic clock comparisons. *Phys. Rev. Lett.* **126**, 011102 (2021).
47. Kobayashi, J., Ogino, A. & Inouye, S. Measurement of the variation of electron-to-proton mass ratio using ultracold molecules produced from laser-cooled atoms. *Nat. Commun.* **10**, 3771 (2019).
48. Patra, S. et al. Proton-electron mass ratio from laser spectroscopy of HD^+ at the part-per-trillion level. *Science* **369**, 1238–1241 (2020).
49. Alighanbari, S., Giri, G. S., Constantin, F. L., Korobov, V. I. & Schiller, S. Precise test of quantum electrodynamics and determination of fundamental constants with HD^+ ions. *Nature* **581**, 152–158 (2020).
50. Germann, M. et al. Three-body QED test and fifth-force constraint from vibrations and rotations of HD^+. *Phys. Rev. Res.* **3**, 022028 (2021).
51. Heacock, B. et al. Pendellösung interferometry probes the neutron charge radius, lattice dynamics, and fifth forces. *Science* **373**, 1239–1243 (2021).
52. Tiberi, E., Borkowski, M., Iritani, B. Moszynski, R. & Zelevinsky, T. Searching for New Fundamental Interactions via Isotopic Shifts in Molecular Lattice Clocks. Preprint at https://arxiv.org/abs/2403.07097 (2024).
53. Wineland, D. J., Bollinger, J. J., Itano, W. M., Moore, F. L. & Heinzen, D. J. Spin squeezing and reduced quantum noise in spectroscopy. *Phys. Rev. A* **46**, 6797–6800 (1992).
54. Wineland, D. J., Bollinger, J. J., Itano, W. M. & Heinzen, D. J. Squeezed atomic states and projection noise in spectroscopy. *Phys. Rev. A* **50**, 67–88 (1994).
55. Ma, J., Wang, X., Sun, C. P. & Nori, F. Quantum spin squeezing. *Phys. Rep.* **509**, 89–165 (2011).
56. Kitagawa, M. & Ueda, M. Squeezed spin states. *Phys. Rev. A* **47**, 5138–5143 (1993).
57. Ritsch, H., Domokos, P., Brennecke, F. & Esslinger, T. Cold atoms in cavity-generated dynamical optical potentials. *Rev. Mod. Phys.* **85**, 553–601 (2013).
58. Hosten, O., Engelsen, N. J., Krishnakumar, R. & Kasevich, M. A. Measurement noise 100 times lower than the quantum-projection limit using entangled atoms. *Nature* **529**, 505–508 (2016).
59. Cox, K. C., Greve, G. P., Weiner, J. M. & Thompson, J. K. Deterministic squeezed states with collective measurements and feedback. *Phys. Rev. Lett.* **116**, 093602 (2016).
60. Pedrozo-Peñafiel, E. et al. Entanglement on an optical atomic-clock transition. *Nature* **588**, 414–418 (2020).
61. Pezzè, L., Smerzi, A., Oberthaler, M. K., Schmied, R. & Treutlein, P. Quantum metrology with nonclassical states of atomic ensembles. *Rev. Mod. Phys.* **90**, 035005 (2018).
62. Perlin, M. A., Qu, C. & Rey, A. M. Spin squeezing with short-range spin-exchange interactions. *Phys. Rev. Lett.* **125**, 223401 (2020).
63. Bilitewski, T. et al. Dynamical generation of spin squeezing in ultracold dipolar molecules. *Phys. Rev. Lett.* **126**, 113401 (2021).
64. Yan, B. et al. Observation of dipolar spin-exchange interactions with lattice-confined polar molecules. *Nature* **501**, 521–525 (2013).
65. Hazzard, K. R. A. et al. Many-body dynamics of dipolar molecules in an optical lattice. *Phys. Rev. Lett.* **113**, 195302 (2014).
66. Christakis, L. et al. Probing site-resolved correlations in a spin system of ultracold molecules. *Nature* **614**, 64–69 (2023).
67. Holland, C. M., Lu, Y. & Cheuk, L. W. On-demand entanglement of molecules in a reconfigurable optical tweezer array. *Science* **382**, 1143–1147 (2023).
68. Bao, Y. et al. Dipolar spin-exchange and entanglement between molecules in an optical tweezer array. *Science* **382**, 1138–1143 (2023).
69. Bornet, G. et al. Scalable spin squeezing in a dipolar Rydberg atom array. *Nature* **621**, 728–733 (2023).
70. Eckner, W. J. et al. Realizing spin squeezing with Rydberg interactions in a programmable optical clock. *Nature* **621**, 734–739 (2023).
71. Franke, J. et al. Quantum-enhanced sensing on optical transitions through finite-range interactions. *Nature* **621**, 740–745 (2023).
72. Hines, J. A. et al. Spin squeezing by Rydberg dressing in an array of atomic ensembles. *Phys. Rev. Lett.* **131**, 063401 (2023).
73. Gorshkov, A. V. et al. Tunable superfluidity and quantum magnetism with ultracold polar molecules. *Phys. Rev. Lett.* **107**, 115301 (2011).
74. Hazzard, K. R. A., Manmana, S. R., Foss-Feig, M. & Rey, A. M. Far-from-equilibrium quantum magnetism with ultracold polar molecules. *Phys. Rev. Lett.* **110**, 075301 (2013).
75. Wellnitz, D., Mamaev, M., Bilitewski, T. & Rey, A. M. Spin squeezing with itinerant dipoles: a case for shallow lattices. *Phys. Rev. Res.* **6**, 012025 (2024).
76. Gorshkov, A. V. et al. Quantum magnetism with polar alkali-metal dimers. *Phys. Rev. A* **84**, 033619 (2011).
77. Tscherbul, T. V., Ye, J. & Rey, A. M. Robust nuclear spin entanglement via dipolar interactions in polar molecules. *Phys. Rev. Lett.* **130**, 143002 (2023).
78. Hermsmeier, R., Rey, A. M. & Tscherbul, T. V. Magnetically tunable electric dipolar interactions of ultracold polar molecules in the quantum ergodic regime. Preprint at https://arxiv.org/abs/2401.04902 (2024).

79. Zhang, C., Yu, P., Jadbabaie, A. & Hutzler, N. R. Quantum-enhanced metrology for molecular symmetry violation using decoherence-free subspaces. *Phys. Rev. Lett.* **131**, 193602 (2023).
80. Garcia Ruiz, R. F. et al. Spectroscopy of short-lived radioactive molecules. *Nature* **581**, 396–400 (2020).
81. Piskorski, J., Patterson, D., Eibenberger, S. & Doyle, J. M. Cooling, spectroscopy and non-sticking of *trans*-stilbene and Nile Red. *ChemPhysChem* **15**, 3800–3804 (2014).
82. Patterson, D. & Doyle, J. M. Sensitive chiral analysis via microwave three-wave mixing. *Phys. Rev. Lett.* **111**, 023008 (2013).
83. Norrgard, E. B., Eckel, S. P., Holloway, C. L. & Shirley, E. L. Quantum blackbody thermometry. *New J. Phys.* **23**, 033037 (2021).
84. Arvanitaki, A., Dimopoulos, S. & Van Tilburg, K. Resonant absorption of bosonic dark matter in molecules. *Phys. Rev. X* **8**, 041001 (2018).
85. Eibenberger, S., Doyle, J. & Patterson, D. Enantiomer-specific state transfer of chiral molecules. *Phys. Rev. Lett.* **118**, 123002 (2017).
86. Doyle, J. M., Lasner, Z. D. & Augenbraun, B. L. High sensitivity chiral detection in the gas phase via microwave spectroscopy and the possible frontier of ultracold chiral molecules. *Chiral Matter* **167**, 195–207 (2023).
87. Quack, M., Stohner, J. & Willeke, M. High-resolution spectroscopic studies and theory of parity violation in chiral molecules. *Annu. Rev. Phys. Chem.* **59**, 741–769 (2008).
88. Cornish, S. L., Tarbutt, M. R. & Hazzard, K. R. A. Quantum computation and quantum simulation with ultracold molecules. *Nat. Phys.* https://doi.org/10.1038/s41567-024-02453-9 (2024).
89. Baron, J. et al. Methods, analysis, and the treatment of systematic errors for the electron electric dipole moment search in thorium monoxide. *New J. Phys.* **19**, 073029 (2017).
90. Fitch, N. J., Lim, J., Hinds, E. A., Sauer, B. E. & Tarbutt, M. R. Methods for measuring the electron's electric dipole moment using ultracold YbF molecules. *Quantum Sci. Technol.* **6**, 014006 (2021).
91. Kozyryev, I. & Hutzler, N. R. Precision measurement of time-reversal symmetry violation with laser-cooled polyatomic molecules. *Phys. Rev. Lett.* **119**, 133002 (2017).
92. Takahashi, Y., Zhang, C., Jadbabaie, A. & Hutzler, N. R. Engineering field-insensitive molecular clock transitions for symmetry violation searches. *Phys. Rev. Lett.* **131**, 183003 (2023).
93. Anderegg, L. et al. Quantum control of trapped polyatomic molecules for eEDM searches. *Science* **382**, 665–668 (2023).
94. Fleig, T. & DeMille, D. Theoretical aspects of radium-containing molecules amenable to assembly from laser-cooled atoms for new physics searches. *New J. Phys.* **23**, 113039 (2021).
95. Kłos, J., Li, H., Tiesinga, E. & Kotochigova, S. Prospects for assembling ultracold radioactive molecules from laser-cooled atoms. *New J. Phys.* **24**, 025005 (2022).
96. Singh, J. T. A new concept for searching for time-reversal symmetry violation using Pa-229 ions trapped in optical crystals. *Hyperfine Interact.* **240**, 29 (2019).
97. Flambaum, V. V. & Dzuba, V. A. Electric dipole moments of atoms and molecules produced by enhanced nuclear Schiff moments. *Phys. Rev. A* **101**, 042504 (2020).
98. Ramachandran, H. D. & Vutha, A. C. Nuclear *T*-violation search using octopole-deformed nuclei in a crystal. *Phys. Rev. A* **108**, 012819 (2023).
99. Kozlov, M. G. & Derevianko, A. Proposal for a sensitive search for the electric dipole moment of the electron with matrix-isolated radicals. *Phys. Rev. Lett.* **97**, 063001 (2006).
100. Li, S. J., Ramachandran, H. D., Anderson, R. & Vutha, A. C. Optical control of BaF molecules trapped in neon ice. *New J. Phys.* **25**, 082001 (2023).

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Competing interests

The authors declare no competing interests.

Additional information

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